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TITLE ALUMINA/SILICA MULTILAYER COATINGS FOR EXCIMER LASERS

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Alumina/Silica Multilayer Coatings for Excimer Lasers*

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The coating parameter that most strongly influences optical damage resistance is the choice of materials used to produce a component. During the course of testing of hundreds of ultraviolet reflectors involving 12 different material combinations, multilayer stacks of Al_2O_3/SiO_2 have demonstrated a superior ability to resist laser-induced damage. Further, damage thresholds for these coatings are at least twice as high as for reflectors composed of other materials: In particular, thresholds of 6 J/cm^2 at 248 nm (15 ns) and 12 J/cm^2 at 351 nm (12 ns) have been measured. Comparative results are presented for a variety of materials at both wavelengths as are preliminary results for alumina-based antireflective coatings.

Key words: Al_2O_3; coating materials; excimer optics; laser-induced damage; multilayer dielectric reflectors; ultraviolet reflectors.

1. Introduction

In this work we report on an extensive survey of materials and vendors for multilayer dielectric reflectors at excimer wavelengths. As a result of this study we provide a ranking of material combinations for ultraviolet reflectors with alumina/silica demonstrating the best performance. In an attempt to explain this ranking, two theoretical models are invoked. Although based upon distinctly different physical phenomena, both demonstrate how Al_2O_3 could be more damage resistant than other common materials and, interestingly, both models predict even better performance for BeO.

2. Test Conditions

Laser pulse lengths at 248 and 351 nm were 15 and 12 ns FWHM, respectively, and the pulse repetition frequency was 35 pps. Both sets of results were generated with a nominally 0.5 mm mean spot diameter; however, a spotsize-independent method of measuring the damage threshold [1] was employed. Briefly, damage threshold was defined as the zero-percent intercept of a damage probability curve, or alternately, as the highest fluence at which damage could not be produced. Damage consisted of physical disruption of the coating which generally began within the first few shots and which frequently evolved, during successive shots, from micron-size pits to a complete failure of the irradiated area. Finally, all testing was of the n-on-m variety wherein m sites were tested at each fluence (m=10) and each nondamaging site was irradiated for n shots (n=140).

3. Results and Discussion

In figure 1 are summarized test data for both 248 nm and 351 nm reflectors with each point representing a single coating run. Excepting the aluminum reflectors, which are shown for purposes of comparison, the coating designs were all of the type

$$S\ H(LH)^nLL$$

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where S is a fused silica substrate, H and L are quarterwave layers of the high- and low-index materials, and n is the number of layer pairs deposited in order to achieve high reflectance. Some general comments on these results are summarized below.

- Thresholds for various material combinations range over nearly two orders of magnitude and, while it is probable that some rearrangement will occur in material rankings as testing continues, only Al_2O_3/SiO_2 has demonstrated 248-nm performance at levels in excess of 4 J/cm^2.

- Assuming, as discussed in a later section, that the high-index material controls damage resistance of a material pair, a question is raised regarding the poor performance of alumina with either NaF or Na_3AlF_6, especially when both were produced by vendors who had been successful with alumina/silica. In the case of the former, the reason is almost certainly related to the fogged condition of the reflectors as delivered. In the alumina/cryolite, however, no problem was indicated by physical appearance or optical performance of the coatings. It was later found that the vendor had used a different coating chamber than that used for alumina/silica depositions, but in the absence of any additional details, no conclusion can be drawn.

- Implicit in figure 1 is a factor-of-two increase in 351-nm thresholds over those for the same materials at 248 nm. Assuming a power-law wavelength dependence, this translates to a threshold scaling of λ^2.

- The reflectors of figure 1 were provided by over fifteen vendors. Alumina/silica samples were purchased from seven vendors; of these seven, four have delivered parts with thresholds over 4 J/cm^2 at 248 nm, and/or over 8 J/cm^2 at 351 nm. These vendors* are:

  Airtron Optical and Magnetic Components
  Broomer Research Corporation
  Coherent Optics Division
  Spectra Physics, Inc. - Optics Division

- Examination of figure 1 reveals, qualitatively, an inverse relationship between threshold and index of refraction of the high-index component. A practical consequence of this is that, for damage-resistant components, system designers will be constrained to use materials with a low index ratio.' The result is shown (fig. 2) in spectral transmittance curves for reflectors of Sc_2O_3/SiO_2 and Al_2O_3/SiO_2. The sample using scandia (n=1.90) achieves a good broadband reflectance with only 23 layers, while the alumina (n=1.65) version requires 49 layers—resulting in the narrow band—to achieve a somewhat lower reflectance.

Although the emphasis thus far has been on reflectors, some preliminary results have been obtained for antireflection coatings as well. Table 1 contains a summary of these results showing that, for an appropriate design, AR thresholds can be as high as for reflectors composed of the same materials.

Table 1. Alumina-Based Antireflection Coatings

Design[a]	Threshold (J/cm^2) 248 nm, 15 ns
S HL	3.4
S LLHL	6.0
S L'L'HL	5.1
S L'L'HL'	4.9

[a] S = substrate (Suprasil 2), H = Al_2O_3, L = SiO_2, L' = MgF_2. All designs had R < (0.5%).

*This list is not the result of a comprehensive vendor's survey, nor does it constitute an endorsement by the University of California, the Los Alamos National Laboratory, or the authors.

4. Modeling the Results

Empirical studies such as the one previously described frequently lose their significance without a suitable physical argument to explain the results. It is fortunate that, in this case, two models exist which potentially offer such an explanation. Unfortunately, at this point neither theory nor experiment is sufficiently mature to allow a conclusion about which model, if either, is correct.

4.1. Damage as an Avalanche Breakdown Process

The first of these two models was presented at the Boulder Damage Symposium in 1975 [2]. It is an avalanche breakdown argument which concludes that the first electrons in the avalanche process are liberated when the local rms electric field in the coating reaches a value proportional to the quantity N/n^2-1, where N is the atomic number density and n is the index of refraction of the material being damaged. It was soon realized, however, that when the proportionality constants are included—the complete expression for the threshold electric field is

$$\frac{N}{n^2-1}\,\frac{q_e}{\varepsilon_o}\,x_{cr}\,\sqrt{10^{-5}}$$

where x_{cr} is the critical electron displacement of about 2 Å—the result is a threshold prediction which is highly optimistic. For the present results, the prediction is optimistic by two orders of magnitude in fluence theshold, or by a factor of ten in terms of field strength. Nevertheless, by ignoring for now the magnitude discrepancy and simply considering the proportionality, very good agreement is found between the material ranking predicted by N/n^2-1 and that observed experimentally (fig. 3).

At this point, a digression is necessary to discuss the assumptions that were made in the construction of figure 3.

- Damage occurs in the high-index component of a multilayer. This follows from the generally accepted argument that high-index materials are more readily damaged than materials with a low index.

- Only the performance envelope of figure 1 is used in the plot of figure 3. This assumes that the envelope represents optimum performance for each material and in addition allows that less damage resistant coatings can be made from any material.

- The factor needed to account for magnitude differences between theory and experiment is approximately the same for all materials tested. This implies that the data in figure 3 should lie on a straight line and that only the slope of the line is in question.

- The linear regression fit in figure 3 uses the origin and all points except three. ZrO_2 and PbF_2 were excluded from the fit because both damaged in a non-normal mode that was indicative of a uniform absorption process—not surprising for these materials at 248 nm. BeO was also excluded but for a different reason: It is assumed that the single coating run evaluated here was not representative of optimized BeO. More on BeO appears at the end of this section.

Returning to the subject of the predicted magnitude of the threshold electric field: We postulate that electric field enhancement at cracks or voids in the coating is responsible for the factor-of-ten discrepancy. It is well known that coatings possess a columnar structure and it is not unreasonable to expect that within this structure exist localized geometric imperfections of an appropriate size. Calculations of electric field enhancement at defects of an appropriate size are available [3], although the enhancement magnitude is far lower than a factor of ten. This subject will be revisited in a future paper.

As mentioned previously, it appears that BeO in figure 3 is not performing as expected. If this is the case continued development work should lead to 248-nm thresholds of about 10 J/cm² for reflectors based on BeO. Work is currently underway to test this hypothesis.

4.2. Damage as a Thermal Process

Another model has recently been proposed [4] in which, instead of being geometric features, the defects are strongly absorbing spherical inclusions (later versions of this model generalize the inclusion shape). The theory predicts that damage occurs when the inclusion/host system reaches some critical temperature and that this temperature is related to the ability of the host to conduct heat away from the defect site. Numerically, the damage threshold fluence should be proportional (for constant pulselength) to $(\rho C_p K)^{1/2}$, where the quantitites represent the density, specific heat, and thermal conductivity, respectively of the host material.

Figure 4 is a plot of various oxide damage thresholds versus the thermal properties of the materials in bulk form. Absolute threshold predictions are not possible due to a general lack of thermal properties of thin films, and because the critical temperature is unknown. As a result of these uncertainties, the credibility of figure 4 is in question. It is shown here because, as measured, good performance is indicated for alumina, and because a completely different physical model has again predicted even better performance for BeO. If the fitted line is correct, 248 nm thresholds in excess of 15 J/cm^2 could be expected for BeO-based reflectors.

It should be noted in closing that preliminary evidence exists [5] which indicates that BeO is more damage resistant than Al_2O_3 in the ultraviolet.

5. Conclusions

We have presented the results of a large survey of vendors and materials for ultraviolet reflectors. We find that Al_2O_3/SiO_2 is the most damage resistant material combination, but that, based upon either of two theoretical models, BeO may prove to be a superior high-index material.

6. References

[1] Foltyn, S. R. Spotsize Effects in Laser Damage Testing. Nat. Bur. Stand. (U.S.) Spec. Publ. 669; 1982, 368 p.

[2] Bettis, J. R.; House, R. A.; Guenther, A. H.; Austin, R. The Importance of Refractive Index, Number Density, and Surface Roughness in the Laser-Induced Damage of Thin Films and Bare Surfaces. Nat. Bur. Stand (U.S.) Spec. Publ. 435; 1975. 289 p.

[3] Bloembergen, N. Role of Cracks, Pores, and Absorbing Inclusions on Laser-Induced Damage Threshold at Surfaces of Transparent Dielectrics. Appl. Opt. 12(4):661-664; 1973 April.

[4] Lange, M. R.; McIver, J. K.; Guenther, A. H.; Walker, T. W. Pulsed Laser Induced Damage of an Optical Material with a Spherical Inclusion: Influence of the Thermal Properties of the Materials. Nat. Bur. Stand. (U.S.) Spec. Publ. 669; 1982. 380 p.

[5] Ebert, J. Activated Reactive Evaporation. Seddon, R. I., ed. Proceedings of Optical Thin Films meeting; 1982 January 26-27; Los Angeles, California. SPIE Vol. 325; 1982. 29-38.

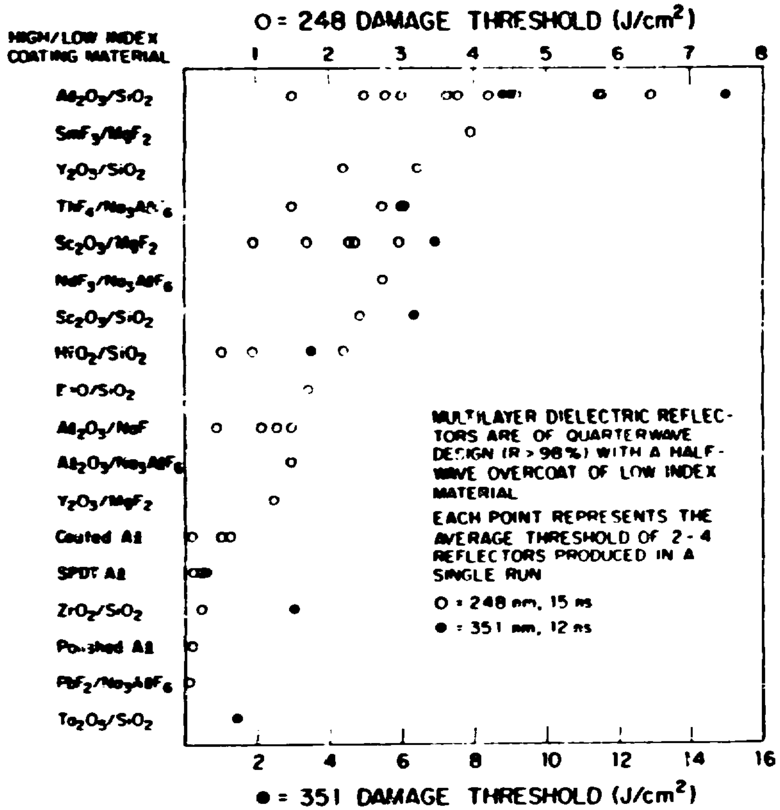

Figure 1. Damage thresholds for various reflector material combinations. Each point represents the average threshold for a single coating run. Dielectric reflectors were of all-quarterwave design with a halfwave overcoat.

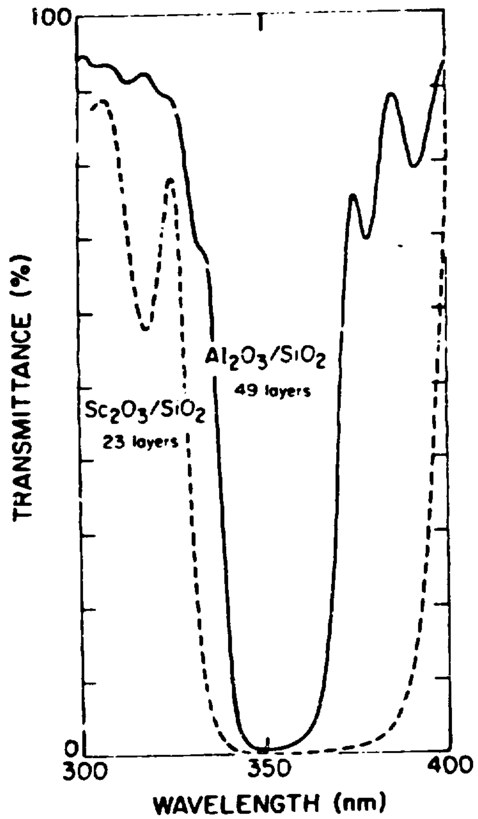

Figure 2. Spectral transmittance curves for two 351 nm reflectors. Although alumina/silica offers the highest damage resistance, it suffers from a low index ratio and a corresponding narrow bandwidth.

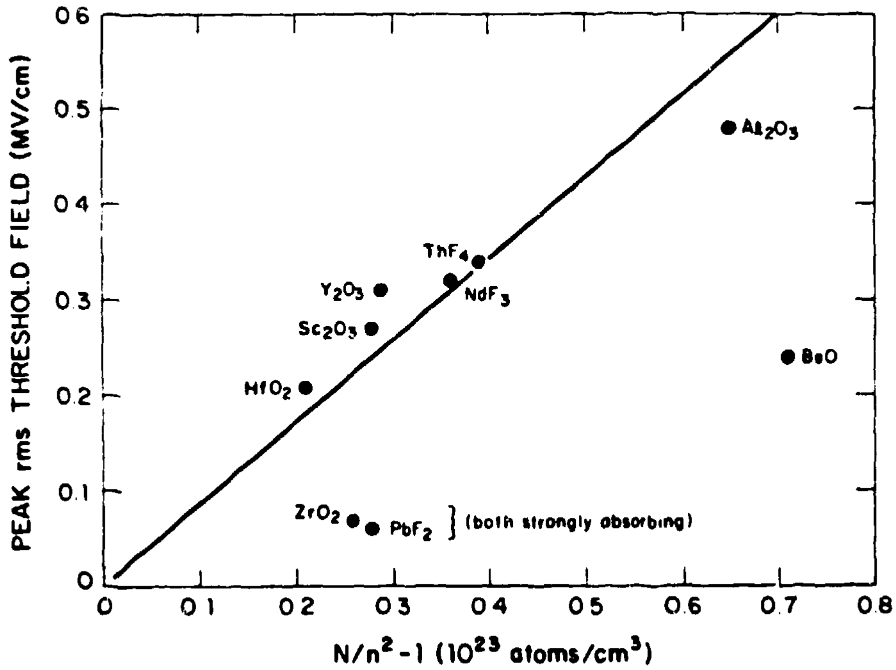

Figure 3. Measured threshold electric fields at 248 nm, 15 ns plotted against N/n^2-1 after reference [2]. While ZrO_2 and PbF_2 absorb strongly at this wavelength, it is postulated that BeO could approach the fitted line with continued optimization.

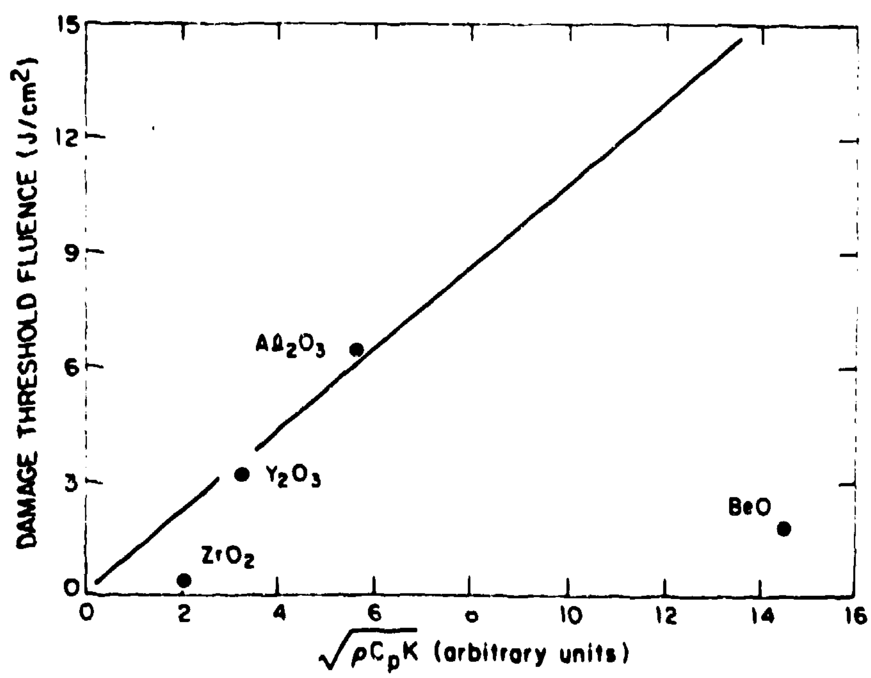

Figure 4. Measured thresholds plotted against $(\rho C_p K)$ after reference [4]. Although lacking thermal data for thin films, this analysis also predicts very high damage resistance for BeO.